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(54) Fully saturated petroleum resin and hot melt pressure sensitive adhesive formulations utilizing same as a tackifier.

(57) This invention relates to pressure sensitive adhesive formulations which include a saturated midblock thermoplastic elastomer such as a block copolymer of polystyrene and hydrogenated polydiene such as Kraton® G and a fully saturated tackifying resin derived from petroleum feedstock polymerized in the presence of aluminium chloride and having a \bar{M}_n ranging from 400 to 800, a softening point of 40 to 70°C and a glass transition temperature less than 45°C whereby the adhesive formulations have a rolling ball tack of less than 10 cm., a 180° peel of from 1 to 4 pounds per inch, a 90° quick stick of from 0.5 to 2.0 pounds per inch and a Polyken probe tack of higher than 400 g/cm² with a formulation containing 100 to 180 phr of the saturated tackifying resin.

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FULLY SATURATED PETROLEUM RESIN AND HOT MELT PRESSURE SENSITIVE ADHESIVE FORMULATIONS UTILIZING SAME AS TACKIFIER 0131460

1 This invention relates to a petroleum resin having
2 excellent tackifying properties for a saturated midblock
3 thermoplastic elastomer. More particularly, it relates to
4 a saturated, water white petroleum resin having a number
5 average molecular weight of from 400 to 800, a softening
6 point from 40°C to 70°C and a glass transition temperature
7 of less than 45°C whereby said resin can provide the re-
8 quisite tackification of pressure sensitive adhesives con-
9 taining a block copolymer of styrene and hydrogenated poly-
10 diene and hot melt pressure sensitive adhesive compositions
11 of both the solution cast and hot melt application types
12 containing said resin and said block copolymer.

13 BACKGROUND OF THE INVENTION

14 Broadly speaking, adhesives can be classified into
15 three categories: water based; solvent based and hot melt
16 adhesives. In the case of water based or solvent based
17 adhesives, energy from the outside, in some fashion, is
18 applied to the system to evaporate the water or the solvent
19 in order that an adhesive layer may be formed. For the
20 solvent based types, the requisite evaporation of the sol-
21 vent also introduces the solvent into the environment which
22 both requires energy and can be deleterious to the surround-
23 ing environment. Costly solvent recovery equipment is then
24 required to control pollution. It is for these reasons that
25 hot melt pressure sensitive adhesives (hereinafter called
26 HMPSA) are highly desirable since the elimination of water
27 and solvent leads to a lower energy requirement to form the
28 adhesive layer with hot melt systems.

29 The key requirement for a HMPSA is that it should
30 have good cohesive and tackifying properties at ambient
31 conditions. The adhesive should also have good flow char-
32 acteristics if it is to be used in the bulk state, otherwise
33 it should possess sufficient consistency when dissolved in
34 suitable cosolvents so that it can be coated or sprayed on
35 the surfaces to be bonded or the backing used to form a tape
36 or label. Usually the HMPSA is prepared from a mixture of
37 tackifying resin, polymer and possibly a plasticizer (when

1 required to soften the adhesive and enhance the aggressive
2 tack).

3 Certain commercial block copolymers such as a
4 Shell's Kraton® are widely used as the polymer in HMPSA. The
5 adhesives prepared from blends incorporating these polymers
6 have very good adhesive and strength properties at room
7 temperature and can be processed by conventional melt coat-
8 ing and extrusion techniques because of their good flow
9 characteristics. Because of this excellent combination of
10 properties exhibited by the ABA type of block copolymer when
11 B represents a polydiene or a polyolefin block and A repre-
12 sents a polystyrene block, e.g. a thermoplastic elastomer,
13 the use of such polymers for various HMPSA applications is
14 growing in the industry.

15 However, these referenced block copolymers when
16 used in combination with known tackifying resins for HMPSA
17 blends have a serious drawback when blended with a major
18 proportion of resin. Under this condition in order to
19 realize the tackifying properties of the petroleum resin
20 component of the blend, it is necessary to incorporate
21 substantial levels of a plasticizer, usually a naphthenic
22 oil or a liquid resin whereby the aggressiveness of tack and
23 other requisite properties of these tertiary blends can be
24 controlled. The use of plasticizers results in a number of
25 deleterious effects on the using of HMPSA compositions in-
26 cluding long term degradation of adhesion.

27 The petroleum resins generally used as tackifiers
28 for Kraton® type block copolymers include those produced
29 according to the teachings of U.S. Patent 3,577,398 wherein
30 the polymeric resinous composition is obtained from the
31 polymerization of a feedstock containing a mixture of pi-
32 perylene, i.e. pentadiene-1,3 and 2-methyl-2-butene in the
33 presence of anhydrous particulate aluminum chloride and a
34 hydrocarbon diluent. The resinous products are reported as
35 having softening points of about 80°C to about 110°C (col.
36 2 line 62) although upon steam stripping the softening
37 points are raised to a range of about 90°C to about 110°C

1 (col. 2, lines 69-70). Unfortunately, as indicated above,
2 the use of a major proportion of such a resin as a tackifier
3 for Kraton® type block copolymers requires from 5 to 30
4 weight percent of plasticizer in the tertiary blend to
5 provide a useful pressure sensitive adhesive composition.
6 Even more critical is the general unsuitability of such a
7 tackifying resin for water-white saturated midblock thermo-
8 plastic elastomers since these resins are colored (not water-
9 white) and have detrimental thermal and ultra-violet
10 light stability.

11 It is an object of this invention to provide an
12 improved petroleum resin for tackification of pressure sen-
13 sitive adhesive blends containing a saturated mid block
14 thermoplastic elastomer.

15 STATEMENT OF THE INVENTION

16 It has been discovered that the polymerization of
17 a C₅ fraction feedstock having a major proportion of a
18 mixture of piperylene and 2-methyl butenes with a total
19 diolefin to monoolefin weight ratio of about 0.47, a total
20 diolefin to 2-methyl butenes weight ratio of about 0.9 and
21 a total cyclic diolefin to monoolefin ratio of about 0.02 in
22 the presence of 1.5 weight percent particulate aluminum
23 chloride produces a petroleum resin having a weight average
24 molecular weight (\bar{M}_w) of 800 to 960, a number average mole-
25 cular weight (\bar{M}_n) of 500 to 600 and a (\bar{M}_w)/(\bar{M}_n) ratio of at
26 least 1.3, which upon hydrogenation to saturation provided
27 a water-white resin having a softening point of about 57°C
28 and a glass transition temperature of about 20°C which in a
29 binary blend with a ABA block copolymer of polystyrene
30 hydrogenated polydiene-polystyrene provided a useful HMPSA
31 composition. Both (\bar{M}_w) and (\bar{M}_n) are measured by gel per-
32 mation chromatography using a polyisobutylene standard and
33 the glass transition temperature (T_g) is a thermal measure-
34 ment using a differential scanning calorimeter (DuPont In-
35 struments Model 990).

36 Thus in accordance with this invention, there is
37 provided a fully saturated petroleum resin possessing in its

1 backbone structure a preponderance of substituted cyclo-
2 hexane rings and having a number average molecular weight
3 (\bar{M}_n) of from 400 to 800, a softening point of 40 to 70°C, a
4 glass transition temperature (T_g) of less than 45°C and when
5 admixed at the total weight level of 60% with Kraton® G 1657,
6 a saturated midblock thermoplastic elastomer, provides a
7 HMPSA having a rolling ball tack of less than 10 cm and a
8 Polyken probe tack greater than 400 gm/cm².

9 The petroleum resin of this invention makes pos-
10 sible in a binary blend a pressure sensitive adhesive formu-
11 lation which comprises (a) a saturated mid-block thermo-
12 plastic elastomer preferably composed of polystyrene blocks
13 and a hydrogenated polydiene block and (b) a fully saturated
14 petroleum resin possessing in its backbone structure a pre-
15 ponderance of substituted cyclohexane rings and having a
16 number average molecular weight (\bar{M}_n) of from 400 to 800, a
17 softening point of 40 to 70°C, a glass transition temperature
18 (T_g) of less than 45°C and when admixed at the total weight
19 level of 60% with Kraton® G 1657, a saturated midblock
20 thermoplastic elastomer and coated to a thickness of 1.5 mil
21 on a polyester sheet backing, e.g. polyethylene terephtha-
22 late provides a HMPSA having a rolling ball tack of less than
23 10 cm and a Polyken probe tack greater than 400 gm/cm².

24 DETAILED DESCRIPTION OF THE INVENTION

25 In practicing this invention to produce the novel
26 petroleum resin which has been found to provide tackifica-
27 tion in binary blends to saturated midblock thermoplastic
28 elastomers, the feedstock composition, polymerization con-
29 ditions and hydrogenation conditions are critical if one is
30 to obtain the fully saturated resin possessing the requisite
31 molecular weight, softening point and preferred glass tran-
32 sition temperature properties.

33 Feedstock

34 The feedstock is obtained as a C₅-C₆ fraction from
35 steam cracked naphtha or light refinery streams with a
36 boiling range of from 20°C to 80°C and a major proportion of
37 piperylene and usefully 2-methyl butenes as the polymeriza-
38 ble content of said fraction. This C₅-C₆ fraction can be

1 usefully adjusted in a suitable way to provide a weight ratio
2 of total diolefin to monoolefin of from 0.7 to 1.0; however,
3 when other less reactive monoolefins are present in the
4 feedstock this latter ratio range is extended without preju-
5 dice to the product resin to from 0.5 to 1.0.

6 The diolefins as hereabove referred to are those
7 represented by cyclopentadiene, cis and trans 1,3-pentadiene
8 and isoprene. The total monoolefins include both five and
9 six carbon hydrocarbons such as 2-methyl-1-butene, 2-methyl-
10 2-butene, cyclopentene, 1-pentene, cis and trans-2-pentene,
11 trans-4-methyl pentene, 2-methyl pentene and 1-hexene. The
12 total 2-methyl butenes include both isomers of the above
13 butenes. The total diolefins include all five and six carbon
14 hydrocarbons such as 1,3-cyclopentadiene.

15 The above said adjustment of weight ratio between
16 the diolefins and monoolefins or 2-methyl butenes may be
17 effected by any of the customary procedures without any
18 particular restriction, such as, for example, removal of
19 isoprene by precise fractional distillation; adjustment of
20 the concentration of diolefins and monoolefins by extractive
21 or azeotropic distillation; removal of diolefins by thermal
22 dimerization as described in U.S. Patent 4,358,574; and
23 addition of a C₅ fraction which has been deprived of di-
24 olefins (i.e. so called spent C₅ fraction). A typical
25 feedstock composition is set forth hereafter in Example 1.

26 Polymerization

27 In carrying out the polymerization reaction, the
28 hydrocarbon mixture is brought into contact with the an-
29 hydrous aluminum chloride catalyst. Generally, the cata-
30 lyst is used in particulate form. Generally a particle size
31 in the range of from about 5 to about 200 mesh size is used
32 although larger or smaller particles can be used. The amount
33 of catalyst used ranges from 1.0 to 2.0, preferably 1.5,
34 weight percent. The catalyst may be added to the olefinic
35 hydrocarbon mixture or the hydrocarbon mixture may be added
36 to the catalyst. The reaction can be conducted continuously
37 or by batch process techniques generally known to those

1 skilled in the art.

2 The reaction should also be carried out at a
3 pressure of from 10 to 80, preferably 20, psi. Since the
4 number average molecular weight of the product polymerizate
5 is important to the invention, the other reaction parameters
6 of: temperature which ranges from 30°C to 100°C, more use-
7 fully from 50°C to 75°C, preferably about 60°C; and, time of
8 polymerization which ranges from 0.5 to 2.0 hours, prefer-
9 ably 1 hour, should be controlled.

10 The reaction is conveniently carried out in the
11 presence of a diluent because the reaction is usually exo-
12 thermic and the resulting product is viscous. However, with
13 adequate mixing and cooling, the temperature can be con-
14 trolled and reaction conducted with only sufficient diluent
15 to maintain good transfer out of the heat of polymerization.
16 The diluent may be introduced as a component of the feedstock
17 for resinification, e.g. in the example alkanes are intro-
18 duced as a significant (about 35%) component of the pi-
19 perylene rich feedstream. Various other diluents which are
20 inert in that they do not enter into the polymerization
21 reaction may be used. Representative examples of inert
22 diluents are aliphatic hydrocarbons such as pentane, hexane
23 and heptane, aromatic hydrocarbons such as toluene and ben-
24 zene, and unreacted residual hydrocarbons from the reac-
25 tion.

26 Unsaturated Precursor Resins

27 The resulting polymeric product can be considered
28 a synthetic hydrocarbon resin which has in its backbone
29 structure a high degree of substituted 1,4-cyclohexene and
30 cyclohexane type of rings. Such rings come about from the
31 backbitting which occurs when a feedstock containing a large
32 percentage of 1,3-dienes (piperlyenes) is polymerized cati-
33 onically with a Lewis acid such as aluminum chloride and
34 utilized 2-methylbutenes for chain transfer or termination.
35 The polymerizate product is characterized by a number aver-
36 age molecular weight (M_n) of from 400 to 800, and softening
37 point ranging from 75-95°C as measured by ASTM E-28.

Hydrogenation

The hydrogenation can be any conventional means which does not affect the molecular weight of the precursor resin, as for example by hydrogenation over a massive nickel catalyst at 200 to 1200 psig hydrogen pressure at 230°C to 270°C for a suitable period. Hydrogenation of the resin for purposes of this invention is realized when the resin is fully saturated (all the C to C double bonds are hydrogenated) and the softening point and glass transition temperature are within the desired range, i.e. 40 to 70°C and less than 47°C, respectively. This level of hydrogenation also destroys the color bodies present in the unsaturated resin.

All of the hydrogenations of the disclosure were performed in a one gallon stainless steel (316) batch autoclave. The catalyst was Harshaw Ni 3266 E 1/8" extrudates which is a massive Ni catalyst. It was contained in a cylindrical basket made of a heavy wire screen which fits snugly against the inner wall of the reactor and had an annulus slightly larger than the turbine blade agitator which was located in the center of this annulus. The catalyst was activated according to the manufacturer's preferred procedure. The hydrogenation procedure was to charge the reactor with 300 grams of the resin dissolved in enough n-heptane to make 1/2 gallon of solution at room temperature. The reactor was pressured to 50 psig with H₂ and then heated as rapidly as possible to the desired temperature but care was taken to ensure this temperature was never exceeded by more than 3°C. The heating time to 200°C was approximately 2 hours. The reactor was then pressured to 900 psig with hydrogen and maintained at this pressure for the duration of the run which was normally 4 hours.

Saturated Resin Tackifier

The resin of the invention is the fully saturated precursor resin discussed above which upon hydrogenation produces a fully saturated petroleum resin possessing in its backbone structure a preponderance of substituted cyclo-

1 hexane rings and having a number average molecular weight
2 (\bar{M}_n) of from 400 to 800, a softening point of 40 to 70°C, a
3 glass transition temperature (T_g) of less than 45°C and when
4 admixed at the level of 60% with Kraton® G 1657, a saturated
5 midblock thermoplastic elastomer, provides a HMPSA having a
6 rolling ball tack of less than 10 cm and a Polyken probe tack
7 greater than 400 gm/cm². The resin of the invention is an
8 excellent tackifier for saturated mid block thermoplastic
9 elastomers as represented by styrene-ethylene/butene-styrene
10 block polymers such as Shell Chemical Company's Kraton® G
11 1657. The softening points are measured by ASTM E-28.

12 Saturated Midblock Thermoplastic Elastomers

13 The saturated midblock thermoplastic elastomers
14 usefully tackified by the resin of this invention are com-
15 mercially exemplified by ABA type block copolymers of (A)
16 polystyrene and (B) a polydiene which is typically selected
17 from the group consisting of polybutadiene and polyisoprene
18 wherein the unsaturated midblock of either polybutadiene or
19 polyisoprene is hydrogenated to yield a saturated midblock
20 segment. Commercially available hydrogenated block copo-
21 lyomers are manufactured by Shell Chemical Company and sold
22 under the tradename: Kraton® G. An extensive discussion of
23 said block copolymers and formulation of them into adhesives
24 can be found in Rubber Chemistry and Technology, 55, pages
25 208-218 (1981). The block copolymers with the saturated
26 midblock segment have an \bar{M}_n as measured by GPC of about
27 25,000 to about 300,000, more preferably about 30,000 to
28 about 200,000, and most preferably about 50,000 to about
29 150,000.

30 Hot Melt Pressure Sensitive Adhesive Blend

31 The HMPSA is generally a blend of the tackifier
32 petroleum resin described herein in amounts ranging from 100
33 to 180, preferably 140 to 180, optimally about 160 parts of
34 resin with 100 parts of the saturated midblock thermoplastic
35 elastomer formulation generally includes a phenolic anti-
36 oxidant such as Irganox® 1010 in amounts ranging from 1 to
37 5, generally 2, weight percent based on the total amount of

of the resin-copolymer components. The HMPSA blends of the invention can also include other tackifiers such as another petroleum resin, diluents and plasticizers and other elastic polymers in order to make minor adjustments in the adhesive properties of the HMPSA.

As indicated a useful approach to defining the tackifying resin of the invention is to measure the adhesive properties of a blend of 150 weight parts tackifying resin (60% of the total blend) with 100 weight parts of a saturated midblock thermoplastic elastomer such as Kraton® G 1657. Such a blend provides a HMPSA having a rolling ball tack of less than 10 cm., usually 2 to 6 cm. and a Polyken probe tack of greater than 400, preferably at least 600, gm./cm.²

The advantages of the HMPSA formulations of the present invention can be more readily appreciated by reference to the following examples.

EXAMPLE 1

As an Example of this invention a precursor petroleum resin was prepared by polymerizing a C₅ to C₆ fraction having a boiling point of 22°C to 65°C having the composition shown in Table I.

TABLE I

Typical Composition of Hydrocarbon Fraction

| | | |
|--|-----|-----------|
| 2-Methylbutane | 29% | by weight |
| 2-Methyl-1-butene | 4% | " " |
| Isoprene | 1% | " " |
| 2-Methyl-2-Butene | 19% | " " |
| Cyclopentadiene | 1% | " " |
| Cis/trans 1,3-pentadiene | 20% | " " |
| Cyclopentene | 7% | " " |
| Other C ₅ /C ₆ Olefins | 13% | " " |
| Other Alkanes | 7% | " " |
| Total diolefins to 2-methyl butenes (wt. ratio) | = | 0.96 |
| Total cyclic diolefins to monoolefins (wt. ratio) | = | 0.02 |
| Total diolefins to mono- olefins (wt. ratio) | = | 0.51 |

1 Four hundred grams of the fraction shown on Table
2 I were fed into a reactor containing 6.0 g of anhydrous AlCl_3
3 dispersed in 10 ml of benzene and equipped with a stirrer,
4 a condenser and a thermometer. As the addition of the feed
5 to the catalyst started, the temperature of the reaction
6 increased slowly until a maximum temperature of 68°C was
7 achieved. The addition of the feed usually takes place
8 during 30 minutes followed by 30 minutes of further reaction.

9 At the end of the 1 hour reaction time the reaction
10 was quenched by adding 200 ml of distilled water. The total
11 reaction contents were placed in a separatory funnel and
12 washed twice with 2 liters of a 3 to 1 mixture of water to
13 isopropanol.

14 The now clean dilute polymerizate was placed in a
15 two-necked flask and heated under nitrogen to 195°C to
16 volatilize the unreacted olefins and alkanes leaving behind
17 a mixture of oligomers and the hydrocarbon resin. This
18 mixture was further stripped by steam to separate the oli-
19 gomers from the resin. This separation was carried out at
20 250°C until 250 ml of steam condensate were collected in the
21 received flask leaving behind 140g of precursor resin having
22 the properties outlined in Table IA.

23 TABLE IA

24 Typical Composition of Hydrocarbon Fraction

25 Serving as the Precursor Resin

| | |
|---|------|
| 26 Softening Point (ring and ball method | 80°C |
| 27 ASTM E-28) | |
| 28 Cloud Point (20/40/40) (Wax/Resin/EVA) | 60°C |
| 29 Gardner Color (50 wt.% resin in toluene) | 6 |
| 30 \bar{M}_w | 890 |
| 31 \bar{M}_n | 560 |
| 32 \bar{M}_w/\bar{M}_n | 1.6 |
| 33 Tg (glass transition temperature as | 53°C |
| 34 measured by thermo mechanical analysis) | |

300 grams of the precursor resin was dissolved in 1.5 to 2 liters of hexane. The solution was placed in a 1 gallon batch hydrogenation reactor and therein the resin was hydrogenated over a massive Ni catalyst (Harshaw 3266) at 1,000 psig hydrogen pressure at 190°C. After approximately 5 hours at the above conditions, the reactor was drained and the hexane was removed by vacuum/steam stripping. The resulting fully saturated resin was water-white and had the following properties outlined in Table IB.

TABLE IB

Composition of Saturated Resin

| | |
|---|-------|
| Softening Point (ring and ball method ASTM E-28) | 59 °C |
| Cloud Point (20/40/40) | 60°C |
| Gardner Color (50 wt.% resin in toluene) | 1 |
| \bar{M}_w | 890 |
| \bar{M}_n | 560 |
| \bar{M}_w/\bar{M}_n | 1.6 |
| Tg (glass transition temperature as measured thermo mechanical analysis) | 36°C |
| Tg (as measured by differential scanning calorimetry using the referenced DuPont apparatus) | 20°C |

The above data show that the hydrogenation lowered the softening point and Tg of the unsaturated resin and provided a water-white, fully saturated tackifying resin of the invention without affecting the molecular weights.

EXAMPLES 2-5

Four HMPSA formulations were each prepared by dissolving by agitation 100 weight units of Kraton® G block copolymer purchased from Shell Chemical Company, Houston, Texas, 2 weight units of a commercial phenolic antioxidant Irganox® 1010 purchased from Ciba-Geigy Corporation, Summit, New Jersey and the indicated weight units of the saturated resin of the invention for Examples 2-4 and a commercial tackifying resin for Example 5 in enough toluene to make a 50 wt% solution. The adhesive was cast from the test

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- 1 formulation onto a 1.5 mil thick polyethylene terephthalate
- 2 (Mylar®) film to provide a dry adhesive film thickness of 1.5
- 3 mil. The results are shown in Table II.

TABLE II
PRESSURE SENSITIVE ADHESIVE FORMULATION RESULTS

| | Formulation PHR* | Example 2 | Example 3 | Example 4 | Example 5 |
|----|--|-----------|-----------|-----------|-----------|
| 3 | Kraton® G (1657)** | 100 | 100 | 100 | 100 |
| 4 | Tablet Resin | 80 | 120 | 160 | 80*** |
| 5 | Antioxidant | 2 | 2 | 2 | 2 |
| 6 | | | | | |
| 7 | <u>Test Results</u> | | | | |
| 8 | Rolling ball tack, cm | | | | |
| 9 | (PSTC Test #6) | 6 | 8 | 8 | 14 |
| 9 | Initial | | | | |
| 10 | 180OC Peel, lb/inch | | | | |
| 11 | (PSTC #1) | | | | |
| 12 | Initial | 2.6 | 3.2 | 3.8 | 2.6 |
| 13 | 90° Quick Stick, lb/inch | | | | |
| 14 | (PSTC Test #1A) | | | | |
| 15 | Initial | 1.4 | 2.1 | 2.7 | 1.2 |
| 16 | Polyken Tack, g/cm ² | | | | |
| 17 | (ASTMD-2979) | | | | |
| 18 | Initial | 870 | 1380 | 1543 | 650 |
| 19 | * PHR refers to "parts hundred rubber" which herein refers to copolymer as the rubber. | | | | |
| 20 | ** Kraton® G (1657) is sold by Shell Chemical Company of Houston, Texas. | | | | |
| 21 | *** Escorez Resin 5380 sold by Exxon Chemical Americas, Houston, Texas. | | | | |
| 22 | PSTC Tests 1, 1A and 6 are published by Pressure Sensitive Tape Council, Glenview, ILL | | | | |
| 23 | 60025. | | | | |
| 24 | ASTMD-2979 refers to the test specified by American Society for Testing Materials. | | | | |

1 These data clearly show that the tackifying resin
2 of the invention is a much superior tackifier for Kraton® G
3 1657 than a commercially available tackifying resin. At
4 twice the loading, 160 phr vs. 80 phr, the new tackifying
5 resin shows superior properties in all categories. In
6 addition, at 160 phr the viscosity of the formulation is low
7 enough so that no processing oil would be needed for good
8 application rates.

9 Where the HMPSA of the invention requires the
10 incorporation of a solvent to facilitate its coating or
11 spraying onto the surfaces to be bonded it is preferred to
12 utilize naphtha as the solvent to provide a concentration
13 level of 35 to about 55 weight percent of Elastomer and
14 saturated petroleum resin dissolved in said solvent, said
15 weight percent based on the total weight of the solution.

CLAIMS:

1. A fully saturated petroleum resin possessing in its backbone structure a preponderance of substituted cyclohexane rings and having a number average molecular weight (\bar{M}_n) of from 400 to 800, a softening point of 40 to 70°C, a glass transition temperature (T_g) of less than 45°C and when admixed at the total weight level of 60% with Kraton® G 1657, a saturated midblock thermoplastic elastomer, provides a HMPSA having a rolling ball tack of less than 10 cm and a Polyken probe tack greater than 400 gm/cm².

2. The petroleum resin according to claim 1 having a glass transition temperature of from 10°C to 30°C and softening point in the range of 50°C to 65°C.

3. The petroleum resin according to claim 2 wherein said admixture exhibits a rolling ball tack of from 2 to 6 cm. and a Polyken probe tack of at least 600.

4. A hot melt pressure sensitive adhesive (HMPSA) formulation which comprises (a) a saturated mid block thermoplastic elastomer and (b) about 100 to 180, parts by weight per 100 parts of weight of said elastomer of a fully saturated petroleum resin possessing in its backbone structure a preponderance of substituted cyclohexane rings and having a number average molecular weight (\bar{M}_n) of from 400 to 800, a softening point of 40 to 70°C, a glass transition temperature (T_g) of less than 45°C and when admixed at the total weight level of 60% with Kraton® G 1657, a saturated midblock thermoplastic elastomer, provides a HMPSA having a rolling ball tack of less than 10 cm and a Polyken probe tack greater than 400 gm/cm².

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5. A HMPSA formulation according to claim 4 containing up to 2 weight percent of an antioxidant.

6. A HMPSA formulation according to claim 4 wherein said elastomer is a block copolymer of styrene and hydrogenated polydiene.

7. A HMPSA formulation according to claim 6 containing a naphtha solvent, said elastomer and saturated petroleum resin being dissolved in said solvent at a concentration level of 35 to about 55 weight percent based on the total weight of said composition.



European Patent
Office

EUROPEAN SEARCH REPORT

0131460

Application number

EP 84 30 4661

| DOCUMENTS CONSIDERED TO BE RELEVANT | | | |
|---|--|--|--|
| Category | Citation of document with indication, where appropriate, of relevant passages | Relevant to claim | CLASSIFICATION OF THE APPLICATION (Int. Cl. 3) |
| A, D | US-A-3 577 398 (HENRY A. PACE) * abstract; column 3, lines 9-18 * | | C 09 J 3/14 C 08 F 240/00 C 08 F 8/04 |
| A | --- EP-A-0 001 306 (SHELL) * page 2, lines 8-19; page 3, lines 1-6; page 5, line 26 - page 6, line 2 * | | |
| A | --- US-A-3 554 940 (M. ARAKAWA) * abstract * ----- | | |
| The present search report has been drawn up for all claims | | | TECHNICAL FIELDS SEARCHED (Int. Cl. 3) |
| | | | C 09 J C 08 F |
| Place of search THE HAGUE | | Date of completion of the search 15-10-1984 | Examiner GIRARD Y.A. |
| CATEGORY OF CITED DOCUMENTS | | | |
| X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document | | T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document | |

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